

Classical-Field Theory of Electron Waves as a Polarized Radiation Probe of Magnetic Surfaces

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History

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abstract environment, an arithmetical error corrected, and typographical errors corrected, released.

- 2002, March 20th: Version 5.2, written by D. C. Hatton and J. A. C. Bland, released.

Endorsements

The “Acknowledgements” section, and sections 1, 2, 3, 4, 5, and 6, of this document, form a transcript of a talk, given in the “Novel Techniques in Magnetism” meeting of the CMD19CMMP 2002 conference, organized by the European Physical Society and the Institute of Physics. The “Abstract” is that which appeared in the abstracts book for this conference, and on the basis of which the talk was accepted for inclusion in the conference.

Abstract

Recently, there has been a revival [17] of interest [4] in mechanisms for changing the spin polarization of an electron beam on transmission through, or reflection from, a magnetic surface. An understanding of these mechanisms would [17] allow the use of an electron beam as a polarized radiation probe for magnetic characterization, like light in MOKE and neutrons in PNR. Here, a mechanism is described which, unlike simultaneously occurring processes proposed elsewhere [17], polarizes an unpolarized incident beam without recourse to inelastic processes.

A magnetic field leads to a Zeeman term in an electron’s Hamiltonian, which depends on the angle θ between the electron’s spin vector and the magnetic flux. As a result, when an electron wave is incident on the surface of a bulk magnetic material (figure 1,) the wave-number of the transmitted wave depends on θ . When the conditions of continuity of the wave-function, and of its first spatial derivative, at the surface, and conservation of particles, are applied, an electron reflection coefficient is obtained which also depends on θ . Therefore, some polarizations are preferentially reflected, while others are preferentially transmitted. The amplitude reflection and transmission coefficients can readily be converted to intensity coefficients, and averaged over an incoherent superposition of electron waves of different θ , e.g. an unpolarized incident beam. The reflected polarization is

$$P = -\frac{2e\mu_B VB}{3e^2 V^2 + \mu_B^2 B^2}, \quad (1)$$

which can take values

$$-\frac{1}{\sqrt{3}} \leq P \leq \frac{1}{\sqrt{3}}, \quad (2)$$

depending on the balance between V and B .

The analysis can be extended to multi-layers using the theory of Fabry-Perot etalons.

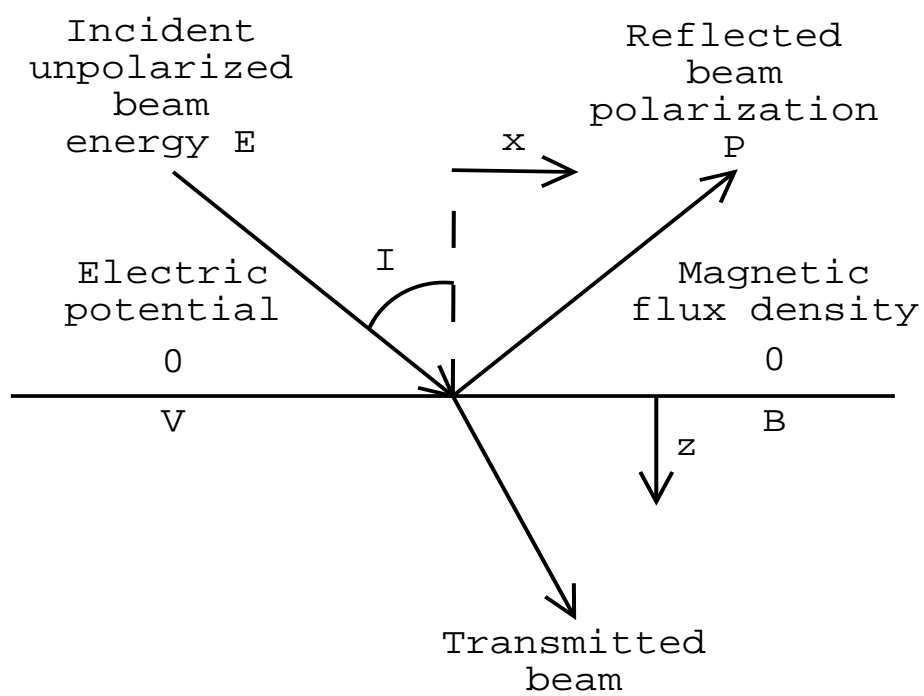


Figure 1: Surface of a Bulk Magnetic Sample

Acknowledgements

Good morning. As you’ve just heard, I’m Dan Hatton, and I’m going to present a classical-field theory of electron waves as a polarized radiation probe of magnetic surfaces.

We’d like to thank the Engineering and Physical Sciences Research Council, in the UK, for paying for this work, and also Thanos Mitrelias, Klaus Peter Kopper, and Peter Bode, our present and past collaborators, in setting up the experiments for which we’re putting forward a theoretical interpretation.

1 Introduction

1.1 Polarized Neutron Reflection

Polarized neutron reflection, or PNR, is [2, 3] an established experimental technique for the measurement of layer-dependent magnetization vector in magnetic multi-layers. A multi-layer structure is [2, 3] modelled as a series of steps in nuclear potential and magnetic flux density. The amplitude reflection coefficient for neutron waves at each step is then calculated by applying the usual [16] boundary conditions to the spin-up and spin-down wave-functions at the step, given the change in wave-vector produced by the potential step. The change in wave-vector depends on the neutron’s spin direction because of the torque exerted upon the neutron magnetic moment, by the magnetic field. Therefore, the spin polarization of the reflected neutron beam, as a function of incident beam energy, provides an indicator of the depth-resolved magnetization profile of the sample.

1.2 Polarized Electron Reflection

Polarized electron reflection and diffraction are [13, 12] also established experimental techniques, for the characterization of magnetic surfaces. The measurement is identical to PNR except for the substitution of electrons for neutrons, and the unavailability [10, 11] of the Stern-Gerlach experiment, either for controlling the incident polarization, or for measuring the reflected polarization. The Stern-Gerlach experiment is [13, 12] typically replaced by a Mott polarimeter [9, 7, 5], for measuring the reflected polarization. Electrons have significant advantages over neutrons for this purpose: an electron beam can be produced using a device roughly equivalent to a light-bulb filament, whereas a neutron beam is typically produced using a nuclear reactor. Also, the magnetic moment of the electron is nearly two thousand times that of the neutron.

Despite the long-standing use of polarized electron reflection as an experimental technique, as far as we’re aware, there has been no attempt to develop a theoretical model of the process, along the lines of that used for PNR, in order to interpret the results in terms of the depth profile of the magnetization in the sample. Our intention here is to produce an analysis of polarized electron reflection similar to that of PNR by Blundell and Bland [2, 3]. If you find the

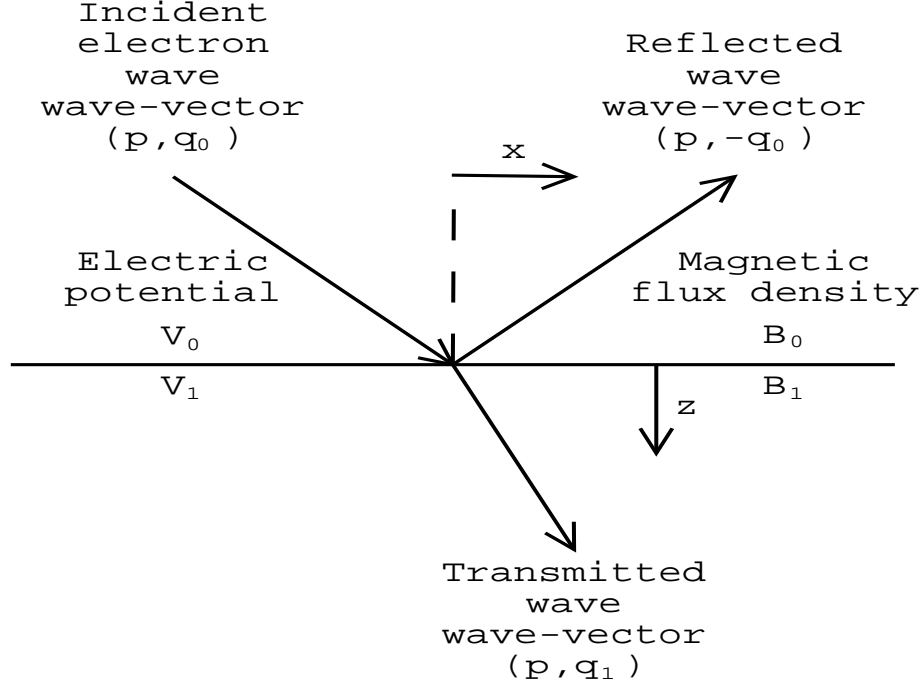


Figure 2: Reflection of an Electron Wave by a Single Step in Electric Potential and Magnetic Flux Density

analysis interesting, a transcript of this talk can be found on the web at this address. The web version also includes more details of the derivations of equations, which are only sketched here. If you can stomach reading all that maths, we'd be grateful for any comments or suggestions. If you can't stomach reading all that maths, I suggest you make yourself difficult to contact around October, so you don't end up being one of the unfortunates who have to examine my thesis.

2 Amplitude Reflection Coefficient for an Electron Pure State, at a Single Step in Electric Potential and Magnetic Flux Density

The first step in the analysis of reflection is to build a potential-theory model of the sample, as a series of steps in electric potential and magnetic flux density (figure 1, figure 2.) Next, we need to discover the amplitude reflection coefficient, for a pure, coherent, electron wave, at a single step (figure 2.) The incident and transmitted electron waves are modelled as plane waves, with well-defined wave-vector components p in the plane of the interface, and q_i perpendicular to the

interface. p must be the same for all the waves, in order to satisfy the boundary condition of continuity of the wave-function at the interface. Strictly, the eigenstates of a Hamiltonian which includes a magnetic field are not plane waves; more about this later (section 5.) The amplitude reflection coefficient is [16] this:

$$r_{01} = \frac{q_0 - q_1}{q_0 + q_1}, \quad (3)$$

or, for a general interface, this one:

$$r_{ij} = \frac{q_i - q_j}{q_i + q_j}. \quad (4)$$

Next, we need to build an expression for the energy of the electrons. There will be kinetic energy terms, along with an electrostatic potential energy, and a term due to the torque, on the electron magnetic moment, in a magnetic field [16]. The form used for this last term assumes a well-defined energy for all directions of the electron spin. Strictly, only certain spin directions are eigenstates of a Hamiltonian which includes a magnetic field; more about this later (section 5.) This leads to this expression

$$q_i = \left(\frac{2m_e E \cos^2 I}{\hbar^2} \right)^{1/2} (1 + x_i)^{1/2}, \quad (5)$$

for the perpendicular wave-vector component, where I represents an angle of incidence, and the potential energy terms are represented by these dimensionless numbers:

$$x_i = y_i + z_i \cos S_i, \quad (6)$$

$$y_i = \frac{eV_i}{E \cos^2 I}, \quad (7)$$

$$z_i = -\frac{e\hbar B_i}{2m_e E \cos^2 I}; \quad (8)$$

I have a big enough ego to call them the Hatton numbers, but I suspect I wouldn't get away with it. S_i is the angle between the electron spin direction and the magnetic flux density in region i , and E is the total energy of the incident electrons, and therefore, by conservation of energy, of all the electrons.

We now use a binomial expansion [6] for the case where the potential energy terms are much smaller than the total electron energy, where the dimensionless numbers we've just devised are small. The magnetic term associated with the Weiss field in a ferromagnet is [15] a few tenths of an electron-volt, and the electrostatic contact potentials in the metals which we study will not be more than a few volts, whereas, in our experimental set-up, the incident electron energies range from a few hundred to a few thousand electron volts, so this approximation seems reasonable. With this expansion, the amplitude reflection coefficient is this:

$$r_{ij} = \frac{1}{4}x_i - \frac{1}{4}x_j - \frac{1}{8}x_i^2 - \frac{1}{8}x_i x_j + \frac{1}{8}x_j^2 + O(\{x_i, x_j\}^3). \quad (9)$$

In the conference talk, I made an error, and had to correct myself, here. Only the second, corrected version appears in this document.

3 Reflection of an Unpolarized Beam from the Surface of a Bulk Magnetic Sample

An unpolarized incident electron beam is [10, 11] an incoherent superposition of pure states representing all directions of the incident spin. The polarization of the reflected beam from any surface is, therefore, given by an average of the polarization over all polarization directions, weighted according to the intensity reflection coefficient for each polarization. This incoherent averaging process (section 7.2) gives this reflected polarization from a bulk surface (figure 1)

$$\begin{aligned} P &= \frac{2y_1 z_1}{3y_1^2 + z_1^2} + O(\{y_1, z_1\}) \\ &= -\frac{4e^2 \hbar m_e V_1 B_1}{12e^2 m_e^2 V_1^2 + e^2 \hbar^2 B_1^2} + O(\{y_1, z_1\}). \end{aligned} \quad (10)$$

Both the term given explicitly, and the next term in the binomial expansion, are in the direction of the magnetic flux density in the bulk material.

The most salient qualitative feature of this polarization formula is that, at high incident electron energies, the reflected polarization is dominated by a non-zero term, which is independent of the incident electron energy, and controlled by the balance between the electrostatic potential and the magnetic flux density, in the sample. This polarization can be as large as $\frac{1}{\sqrt{3}}$ in either direction.

4 Multi-Layer Structures

We propose to extend this analysis to magnetic multi-layer structures, by using the theory of Fabry-Perot etalons, as is [2, 3] already the practice in PNR. There are infinitely many possible paths for reflection from a multi-layer structure, indexed by how many times the electron wave “bounces” within each layer. In the diagram (figure 3,) we can see paths with no bounces, with one bounce, and with two bounces. For a given, pure incident wave, the reflected waves from the various paths are superposed coherently to build the reflected wave, each term in the coherent superposition including an amplitude factor due to the amplitude reflection or transmission coefficient at each interface which it has encountered, and a phase factor due to the path length which it has traversed in the magnetic layers. This will result in a spin-dependent amplitude reflection coefficient for the whole multi-layer system, which will provide the weightings to go into the incoherent superposition over an unpolarized incident beam. This incoherent superposition, as for the bulk sample, will give the reflected polarization. We expect working through the maths for this to be trivial, but time-consuming.

In the conference talk, I made an error, and had to correct myself, here. Only the second, corrected version appears in this document.

5 Comments on This Analysis

I promised to comment on some assumptions in this analysis. Firstly, there’s the matter of modelling the electrons as a plane wave. This is equivalent to ne-

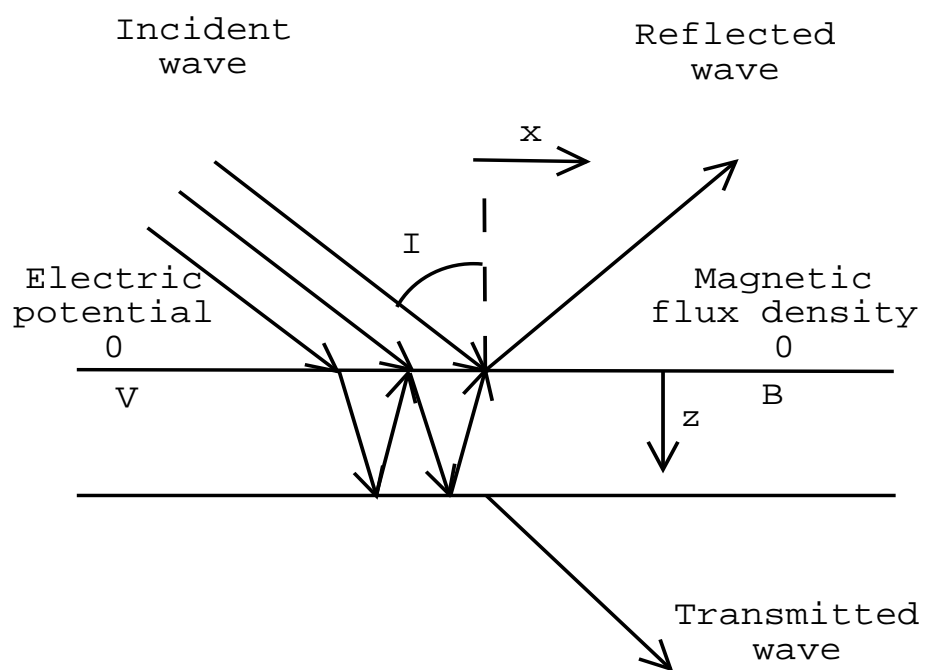


Figure 3: Reflection Paths for an Electron Wave in a Single Magnetic Layer

glecting the deflection of the electrons by the Lorentz force, which means taking the limit of weak magnetic fields; something we’ve done in the binomial expansion anyway. The same convention of neglecting this deflection was adopted by Weber *et al.* [17], when they analysed the spin polarization of transmitted electron waves.

Secondly, there’s the issue of pretending that all electron spin directions are eigen-states of the Hamiltonian. In this we depart from the tradition of analysis of PNR, where matrices are [2, 3] used to represent the Zeeman energy, and the reflection coefficient, without any need for this approximation. We also depart from the work of Weber *et al.* [17] on electron transmission: they regard the Larmor precession, which is a manifestation of the fact that not all spin directions are eigen-states of the Hamiltonian in a magnetic field, as crucial in determining the transmitted polarization. We intend to produce a more “first-principles” model in the near future, which will use the matrix representation of the reflection coefficients, and therefore capture the Larmor precession, and other spin-flip scattering effects. However, we don’t intend to devise this model as a replacement for the one presented here, but as a complement to it. What we’d like to do is subject both models, along with a third, completely classical, reflection model, to experimental data, and use the well-established [14] methods of Bayesian statistics, first to infer the parameters of magnetic flux density, electric potential, and layer thickness, for each model, then to judge the relative confidence which we have in each model.

One reason for not simply abandoning all but the most “first-principles” of the models is given by Anderson [1], who points out that any system, more complicated than a molecule of four atoms or so, is pretty well never in an eigen-state of its Hamiltonian, so the Schrödinger equation doesn’t describe the state of the system. This is because the tunnelling-like processes, which would otherwise collapse the system into an eigen-state of its Hamiltonian, are very slow for complicated systems: often very slow compared with the age of the universe, and certainly very slow compared with the rate of occurrence of measurement-like interactions with the outside world, which collapse the system into eigen-states of operators other than the Hamiltonian. Therefore, it can’t be guaranteed that the model which implements a Schrödinger equation with the most realistic Hamiltonian will always be the most useful in describing the real behaviour of the system.

I might be inclined to add to this a very different argument [8] for not always preferring the most first-principles model, but this isn’t the time or the place for my speculations on mathematical philosophy. Anyone who has a burning desire to hear them can find them via the reference on the slide.

Thirdly, it’s worth commenting on the effect on the polarization of transmitted waves, due to spin-dependent loss of electrons to inelastic processes, which was noted by Weber *et al.* [17]. At first glance, our classical-field analysis appears to be entirely elastic. However, it is capable of assimilating the effect of these processes, which will appear as imaginary parts in the electric potential and magnetic flux density.

6 Conclusions

OK. What have we learnt?

- The spin polarization of the reflected electron beam from a bulk magnetic surface, in the model described, is this:

$$\mathbf{P} = -\frac{4e^2\hbar m_e V_1 \mathbf{B}_1}{12e^2 m_e^2 V_1^2 + e^2 \hbar^2 B_1^2} + O(\{y_1, z_1\}). \quad (11)$$

- The extension of the model to multi-layer systems is likely to be a trivial, but time-consuming, mathematical task.
- Two other, similar models are planned, one of which differs from this by the use of a more “first-principles” treatment of the Zeeman energy, and the other by a fully classical treatment of the reflection process, and
- there is a strong case for retaining all three models, and using Bayesian statistics to compare them in the light of experimental data, rather than discarding all but the most “first-principles” model.

Thank you for listening. I’ll show the slides of references gradually, while I field some questions.

7 Supplementary Mathematical Details

7.1 Amplitude Reflection Coefficient for an Electron Pure State, at a Single Step in Electric Potential and Magnetic Flux Density

The incident and transmitted electron waves are modelled (figure 2) as plane waves, allowing the well-defined wave-vector components p in the plane of the interface, and q_i perpendicular to the interface. p must be the same for all the waves, in order to satisfy the boundary condition of continuity of the wave-function at the interface. The amplitude reflection coefficient is [16]

$$r_{01} = \frac{q_0 - q_1}{q_0 + q_1}, \quad (12)$$

or, for a general interface,

$$r_{ij} = \frac{q_i - q_j}{q_i + q_j}. \quad (13)$$

Next, we need to build an expression for the energy of the electrons. There will be kinetic energy terms, which, in the non-relativistic limit, are

$$\frac{\hbar^2 p^2}{2m_e},$$

This actually failed to happen, except for the first four references: answering the questions kept me too busy to change over the slides. If you’re reading this document because you wanted to know about the references, you’re in luck. They’re here, and numbered in the same way as in the slides.

and

$$\frac{\hbar^2 q_i^2}{2m_e},$$

along with an electrostatic potential energy

$$-eV_i,$$

and a term due to the torque, on the electron magnetic moment, in a magnetic field [16]

$$\frac{e\hbar B_i \cos S_i}{2m_e},$$

where S_i is the angle between the electron spin and the magnetic flux density. The form of this last term assumes a well-defined energy for all values of S_i . Strictly, only certain S_i values are eigen-states of a Hamiltonian which includes a magnetic field; more about this later (section 5.) The total energy is

$$E = \frac{\hbar^2 p^2}{2m_e} + \frac{\hbar^2 q_i^2}{2m_e} - eV_i + \frac{e\hbar B_i \cos S_i}{2m_e}, \quad (14)$$

or, where p is expressed as a fraction $\sin I$ of the total wave-number in the absence of any potential, I being an angle of incidence like that in figure 1,

$$E = E \sin^2 I + \frac{\hbar^2 q_i^2}{2m_e} - eV_i + \frac{e\hbar B_i \cos S_i}{2m_e} \quad (15)$$

$$\begin{aligned} \Rightarrow q_i &= \left(\frac{2m_e E \cos^2 I}{\hbar^2} \right)^{1/2} \left(1 + \frac{eV_i}{E \cos^2 I} - \frac{e\hbar B_i \cos S_i}{2m_e E \cos^2 I} \right)^{1/2} \\ &= \left(\frac{2m_e E \cos^2 I}{\hbar^2} \right)^{1/2} (1 + x_i)^{1/2}, \end{aligned} \quad (16)$$

where $x_i = y_i + z_i \cos S_i$, $y_i = \frac{eV_i}{E \cos^2 I}$, and $z_i = -\frac{e\hbar B_i}{2m_e E \cos^2 I}$.

We now use a binomial expansion [6] for the case where the potential energy terms are much smaller than the total electron energy, where the dimensionless numbers we've just devised are small. The magnetic term associated with the Weiss field in a ferromagnet is [15] a few tenths of an electron-volt, and the electrostatic contact potentials in the metals which we study will not be more than a few volts, whereas, in our experimental set-up, the incident electron energies range from a few hundred to a few thousand electron volts, so this approximation seems reasonable.

$$q_i = \left(\frac{2m_e E \cos^2 I}{\hbar^2} \right)^{1/2} \left(1 + \frac{1}{2}x_i - \frac{1}{8}x_i^2 + O(x_i^3) \right). \quad (17)$$

The amplitude reflection coefficient is, therefore,

$$r_{ij} = \frac{1}{2} \left(\frac{1}{2}x_i - \frac{1}{2}x_j + \frac{1}{8}x_j^2 - \frac{1}{8}x_i^2 + O(\{x_i, x_j\}^3) \right) \left(1 + \frac{1}{4}x_i + \frac{1}{4}x_j - \frac{1}{16}x_i^2 - \frac{1}{16}x_j^2 + O(\{x_i, x_j\}^3) \right)^{-1}$$

$$\begin{aligned}
&= \frac{1}{2} \left(\frac{1}{2}x_i - \frac{1}{2}x_j + \frac{1}{8}x_j^2 - \frac{1}{8}x_i^2 + O(\{x_i, x_j\}^3) \right) \left(1 - \frac{1}{4}x_i - \frac{1}{4}x_j + \frac{1}{8}x_i^2 + \frac{1}{8}x_i x_j + \frac{1}{8}x_j^2 + O(\{x_i, x_j\}^3) \right) \\
&= \frac{1}{2} \left(\frac{1}{2}x_i - \frac{1}{2}x_j - \frac{1}{4}x_i^2 - \frac{1}{4}x_i x_j + \frac{1}{4}x_j^2 + O(\{x_i, x_j\}^3) \right) \\
&= \frac{1}{4}x_i - \frac{1}{4}x_j - \frac{1}{8}x_i^2 - \frac{1}{8}x_i x_j + \frac{1}{8}x_j^2 + O(\{x_i, x_j\}^3).
\end{aligned} \tag{18}$$

7.2 Reflection of an Unpolarized Beam from the Surface of a Bulk Magnetic Sample

An unpolarized incident electron beam is [10, 11] an incoherent superposition of pure states representing all directions of the incident spin. Each such direction can be represented by its spherical polar angle co-ordinates (θ, ϕ) . That is to say, the incident beam contains a flux of electrons

$$F_1 d\theta d\phi = A \sin \theta d\theta d\phi \tag{19}$$

with polarization direction between θ and $\theta + d\theta$, and between ϕ and $\phi + d\phi$. The flux of such electrons in the reflected beam will, therefore, be

$$F_2 d\theta d\phi = |r_{ij}|^2 F_1 d\theta d\phi. \tag{20}$$

The reflection from the surface of a bulk sample is to be modelled as a single reflection, of amplitude reflection coefficient r_{01} , in a situation where V_0 , B_0 , and therefore x_0 , are all zero. In this case,

$$r_{01} = -\frac{1}{4}x_1 + \frac{1}{8}x_1^2 + O(x_1^3), \tag{21}$$

and

$$|r_{01}|^2 = \frac{1}{16}x_1^2 - \frac{1}{16}x_1^3 + O(x_1^4), \tag{22}$$

assuming that r_{01} is real.

If the spherical polar representation (θ_i, ϕ_i) is used for the direction of the magnetic flux density in region i , then

$$\cos S_i = \sin \theta_i \cos \phi_i \sin \theta \cos \phi + \sin \theta_i \sin \phi_i \sin \theta \sin \phi + \cos \theta_i \cos \theta. \tag{23}$$

Therefore,

$$x_i = y_i + z_i (\sin \theta_i \cos \phi_i \sin \theta \cos \phi + \sin \theta_i \sin \phi_i \sin \theta \sin \phi + \cos \theta_i \cos \theta). \tag{24}$$

The polarization of the pure state represented by (θ, ϕ) , in the Cartesian co-ordinate system associated with this spherical polar system, is

$$\mathbf{P}(\theta, \phi) = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta), \tag{25}$$

and the average polarization of the reflected beam is

$$\begin{aligned}
\mathbf{P} &= \frac{\int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} \mathbf{P}(\theta, \phi) F_2 d\theta d\phi}{\int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} F_2 d\theta d\phi} \\
&= \frac{\int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta) |r_{01}|^2 F_1 d\theta d\phi}{\int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} |r_{01}|^2 F_1 d\theta d\phi} \\
&= \frac{\int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta) \left(\frac{1}{16} x_1^2 - \frac{1}{16} x_1^3 + O(x_1^4) \right) \sin \theta d\theta d\phi}{\int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} \left(\frac{1}{16} x_1^2 - \frac{1}{16} x_1^3 + O(x_1^4) \right) \sin \theta d\theta d\phi} \\
&= \frac{(I_2 - I_6, I_3 - I_7, I_4 - I_8) + O(\{y_1, z_1\}^4)}{I_1 - I_5 + O(\{y_1, z_1\}^4)}. \tag{26}
\end{aligned}$$

The crucial integrals are

$$\begin{aligned}
I_1 &= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} x_1^2 \sin \theta d\theta d\phi \\
&= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} (y_1 + z_1 (\sin \theta_1 \cos \phi_1 \sin \theta \cos \phi + \sin \theta_1 \sin \phi_1 \sin \theta \sin \phi + \cos \theta_1 \cos \theta))^2 \sin \theta d\theta d\phi \\
&= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} (y_1^2 \sin \theta \\
&\quad + 2y_1 z_1 (\sin \theta_1 \cos \phi_1 \sin^2 \theta \cos \phi + \sin \theta_1 \sin \phi_1 \sin^2 \theta \sin \phi + \cos \theta_1 \sin \theta \cos \theta) \\
&\quad + z_1^2 (\sin^2 \theta_1 \cos^2 \phi_1 \sin^3 \theta \cos^2 \phi + 2 \sin^2 \theta_1 \sin \phi_1 \cos \phi_1 \sin^3 \theta \sin \phi \cos \phi \\
&\quad + 2 \sin \theta_1 \cos \theta_1 \cos \phi_1 \sin^2 \theta \cos \theta \cos \phi + \sin^2 \theta_1 \sin^2 \phi_1 \sin^3 \theta \sin^2 \phi \\
&\quad + 2 \sin \theta_1 \cos \theta_1 \sin \phi_1 \sin^2 \theta \cos \theta \sin \phi + \cos^2 \theta_1 \sin \theta \cos^2 \theta)) d\theta d\phi \\
&= 4\pi y_1^2 + \frac{4\pi z_1^2}{3}, \tag{27}
\end{aligned}$$

$$\begin{aligned}
I_2 &= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} x_1^2 \sin^2 \theta \cos \phi d\theta d\phi \\
&= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} (y_1 + z_1 (\sin \theta_1 \cos \phi_1 \sin \theta \cos \phi + \sin \theta_1 \sin \phi_1 \sin \theta \sin \phi + \cos \theta_1 \cos \theta))^2 \sin^2 \theta \cos \phi d\theta d\phi \\
&= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} (y_1^2 \sin^2 \theta \cos \phi \\
&\quad + 2y_1 z_1 (\sin \theta_1 \cos \phi_1 \sin^3 \theta \cos^2 \phi + \sin \theta_1 \sin \phi_1 \sin^3 \theta \sin \phi \cos \phi + \cos \theta_1 \sin^2 \theta \cos \theta \cos \phi) \\
&\quad + z_1^2 (\sin^2 \theta_1 \cos^2 \phi_1 \sin^4 \theta \cos^3 \phi + 2 \sin^2 \theta_1 \sin \phi_1 \cos \phi_1 \sin^4 \theta \sin \phi \cos^2 \phi \\
&\quad + 2 \sin \theta_1 \cos \theta_1 \cos \phi_1 \sin^3 \theta \cos \theta \cos^2 \phi + \sin^2 \theta_1 \sin^2 \phi_1 \sin^4 \theta \sin^2 \phi \cos \phi \\
&\quad + 2 \sin \theta_1 \cos \theta_1 \sin \phi_1 \sin^3 \theta \cos \theta \sin \phi \cos \phi + \cos^2 \theta_1 \sin^2 \theta \cos^2 \theta \cos \phi)) d\theta d\phi \\
&= \frac{8\pi y_1 z_1 \sin \theta_1 \cos \phi_1}{3}, \tag{28}
\end{aligned}$$

$$\begin{aligned}
I_3 &= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} x_1^2 \sin^2 \theta \sin \phi d\theta d\phi \\
&= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} (y_1 + z_1 (\sin \theta_1 \cos \phi_1 \sin \theta \cos \phi + \sin \theta_1 \sin \phi_1 \sin \theta \sin \phi + \cos \theta_1 \cos \theta))^2 \sin^2 \theta \sin \phi d\theta d\phi \\
&= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} (y_1^2 \sin^2 \theta \sin \phi \\
&\quad + 2y_1 z_1 (\sin \theta_1 \cos \phi_1 \sin^3 \theta \sin \phi \cos \phi + \sin \theta_1 \sin \phi_1 \sin^3 \theta \sin^2 \phi + \cos \theta_1 \sin^2 \theta \cos \theta \sin \phi) \\
&\quad + z_1^2 (\sin^2 \theta_1 \cos^2 \phi_1 \sin^4 \theta \sin \phi \cos^2 \phi + 2 \sin^2 \theta_1 \sin \phi_1 \cos \phi_1 \sin^4 \theta \sin^2 \phi \cos \phi \\
&\quad + 2 \sin \theta_1 \cos \theta_1 \cos \phi_1 \sin^3 \theta \cos \theta \sin \phi \cos \phi + \sin^2 \theta_1 \sin^2 \phi_1 \sin^4 \theta \sin^3 \phi \\
&\quad + 2 \sin \theta_1 \cos \theta_1 \sin \phi_1 \sin^3 \theta \cos \theta \sin^2 \phi + \cos^2 \theta_1 \sin^2 \theta \cos^2 \theta \sin \phi)) d\theta d\phi \\
&= \frac{8\pi y_1 z_1 \sin \theta_1 \sin \phi_1}{3}, \tag{29}
\end{aligned}$$

$$\begin{aligned}
I_4 &= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} x_1^2 \sin \theta \cos \theta d\theta d\phi \\
&= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} (y_1 + z_1 (\sin \theta_1 \cos \phi_1 \sin \theta \cos \phi + \sin \theta_1 \sin \phi_1 \sin \theta \sin \phi + \cos \theta_1 \cos \theta))^2 \sin \theta \cos \theta d\theta d\phi \\
&= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} (y_1^2 \sin \theta \cos \theta \\
&\quad + 2y_1 z_1 (\sin \theta_1 \cos \phi_1 \sin^2 \theta \cos \theta \cos \phi + \sin \theta_1 \sin \phi_1 \sin^2 \theta \cos \theta \sin \phi + \cos \theta_1 \sin \theta \cos^2 \theta) \\
&\quad + z_1^2 (\sin^2 \theta_1 \cos^2 \phi_1 \sin^3 \theta \cos \theta \cos^2 \phi + 2 \sin^2 \theta_1 \sin \phi_1 \cos \phi_1 \sin^3 \theta \cos \theta \sin \phi \cos \phi \\
&\quad + 2 \sin \theta_1 \cos \theta_1 \cos \phi_1 \sin^2 \theta \cos^2 \theta \cos \phi + \sin^2 \theta_1 \sin^2 \phi_1 \sin^3 \theta \cos \theta \sin^2 \phi \\
&\quad + 2 \sin \theta_1 \cos \theta_1 \sin \phi_1 \sin^2 \theta \cos^2 \theta \sin \phi + \cos^2 \theta_1 \sin \theta \cos^3 \theta)) d\theta d\phi \\
&= \frac{8\pi y_1 z_1 \cos \theta_1}{3}, \tag{30}
\end{aligned}$$

$$\begin{aligned}
I_5 &= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} x_1^3 \sin \theta d\theta d\phi \\
&= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} (y_1 + z_1 (\sin \theta_1 \cos \phi_1 \sin \theta \cos \phi + \sin \theta_1 \sin \phi_1 \sin \theta \sin \phi + \cos \theta_1 \cos \theta))^3 \sin \theta d\theta d\phi \\
&= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} (y_1^3 \sin \theta \\
&\quad + 3y_1^2 z_1 (\sin \theta_1 \cos \phi_1 \sin^2 \theta \cos \phi + \sin \theta_1 \sin \phi_1 \sin^2 \theta \sin \phi + \cos \theta_1 \sin \theta \cos \theta) \\
&\quad + 3y_1 z_1^2 (\sin^2 \theta_1 \cos^2 \phi_1 \sin^3 \theta \cos^2 \phi + 2 \sin^2 \theta_1 \sin \phi_1 \cos \phi_1 \sin^3 \theta \sin \phi \cos \phi \\
&\quad + 2 \sin \theta_1 \cos \theta_1 \cos \phi_1 \sin^2 \theta \cos \theta \cos \phi + \sin^2 \theta_1 \sin^2 \phi_1 \sin^3 \theta \sin^2 \phi \\
&\quad + 2 \sin \theta_1 \cos \theta_1 \sin \phi_1 \sin^2 \theta \cos \theta \sin \phi + \cos^2 \theta_1 \sin \theta \cos^2 \theta) \\
&\quad + z_1^3 (\sin^3 \theta_1 \cos^3 \phi_1 \sin^4 \theta \cos^3 \phi + 3 \sin^3 \theta_1 \sin \phi_1 \cos^2 \phi_1 \sin^4 \theta \sin \phi \cos^2 \phi \\
&\quad + 3 \sin^2 \theta_1 \cos \theta_1 \cos^2 \phi_1 \sin^3 \theta \cos \theta \cos^2 \phi + 3 \sin^3 \theta_1 \sin^2 \phi_1 \cos \phi_1 \sin^4 \theta \sin^2 \phi \cos \phi) \\
&\quad + 3 \sin^2 \theta_1 \cos \theta_1 \cos^2 \phi_1 \sin^3 \theta \cos \theta \cos^2 \phi + 3 \sin^3 \theta_1 \sin^2 \phi_1 \cos \phi_1 \sin^4 \theta \sin^2 \phi \cos \phi) d\theta d\phi
\end{aligned}$$

$$\begin{aligned}
& +6 \sin^2 \theta_1 \cos \theta_1 \sin \phi_1 \cos \phi_1 \sin^3 \theta \cos \theta \sin \phi \cos \phi + 3 \sin \theta_1 \cos^2 \theta_1 \cos \phi_1 \sin^2 \theta \cos^2 \theta \cos \phi \\
& + \sin^3 \theta_1 \sin^3 \phi_1 \sin^4 \theta \sin^3 \phi + 3 \sin^2 \theta_1 \cos \theta_1 \sin^2 \phi_1 \sin^3 \theta \cos \theta \sin^2 \phi \\
& + 3 \sin \theta_1 \cos^2 \theta_1 \sin \phi_1 \sin^2 \theta \cos^2 \theta \sin \phi + \cos^3 \theta_1 \sin \theta \cos^3 \theta)) d\theta d\phi \\
= & 4\pi y_1 z_1^2, \tag{31}
\end{aligned}$$

$$\begin{aligned}
I_6 &= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} x_1^3 \sin^2 \theta \cos \phi d\theta d\phi \\
&= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} (y_1^3 \sin^2 \theta \cos \phi \\
&+ 3y_1^2 z_1 (\sin \theta_1 \cos \phi_1 \sin^3 \theta \cos^2 \phi + \sin \theta_1 \sin \phi_1 \sin^3 \theta \sin \phi \cos \phi + \cos \theta_1 \sin^2 \theta \cos \theta \cos \phi) \\
&+ 3y_1 z_1^2 (\sin^2 \theta_1 \cos^2 \phi_1 \sin^4 \theta \cos^3 \phi + 2 \sin^2 \theta_1 \sin \phi_1 \cos \phi_1 \sin^4 \theta \sin \phi \cos^2 \phi \\
&+ 2 \sin \theta_1 \cos \theta_1 \cos \phi_1 \sin^3 \theta \cos \theta \cos^2 \phi + \sin^2 \theta_1 \sin^2 \phi_1 \sin^4 \theta \sin^2 \phi \cos \phi \\
&+ 2 \sin \theta_1 \cos \theta_1 \sin \phi_1 \sin^3 \theta \cos \theta \sin \phi \cos \phi + \cos^2 \theta_1 \sin^2 \theta \cos^2 \theta \cos \phi) \\
&+ z_1^3 (\sin^3 \theta_1 \cos^3 \phi_1 \sin^5 \theta \cos^4 \phi + 3 \sin^3 \theta_1 \sin \phi_1 \cos^2 \phi_1 \sin^5 \theta \sin \phi \cos^3 \phi \\
&+ 3 \sin^2 \theta_1 \cos \theta_1 \cos^2 \phi_1 \sin^4 \theta \cos \theta \cos^3 \phi + 3 \sin^3 \theta_1 \sin^2 \phi_1 \cos \phi_1 \sin^5 \theta \sin^2 \phi \cos^2 \phi \\
&+ 6 \sin^2 \theta_1 \cos \theta_1 \sin \phi_1 \cos \phi_1 \sin^4 \theta \cos \theta \sin \phi \cos^2 \phi + 3 \sin \theta_1 \cos^2 \theta_1 \cos \phi_1 \sin^3 \theta \cos^2 \theta \cos^2 \phi \\
&+ \sin^3 \theta_1 \sin^3 \phi_1 \sin^5 \theta \sin^3 \phi \cos \phi + 3 \sin^2 \theta_1 \cos \theta_1 \sin^2 \phi_1 \sin^4 \theta \cos \theta \sin^2 \phi \cos \phi \\
&+ 3 \sin \theta_1 \cos^2 \theta_1 \sin \phi_1 \sin^3 \theta \cos^2 \theta \sin \phi \cos \phi + \cos^3 \theta_1 \sin^2 \theta \cos^3 \theta \cos \phi)) d\theta d\phi \\
&= \left(4\pi y_1^2 z_1 + \frac{4\pi z_1^3}{5} \right) \sin \theta_1 \cos \phi_1, \tag{32}
\end{aligned}$$

$$\begin{aligned}
I_7 &= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} x_1^3 \sin^2 \theta \sin \phi d\theta d\phi \\
&= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} (y_1^3 \sin^2 \theta \sin \phi \\
&+ 3y_1^2 z_1 (\sin \theta_1 \cos \phi_1 \sin^3 \theta \sin \phi \cos \phi + \sin \theta_1 \sin \phi_1 \sin^3 \theta \sin^2 \phi + \cos \theta_1 \sin^2 \theta \cos \theta \sin \phi) \\
&+ 3y_1 z_1^2 (\sin^2 \theta_1 \cos^2 \phi_1 \sin^4 \theta \sin \phi \cos^2 \phi + 2 \sin^2 \theta_1 \sin \phi_1 \cos \phi_1 \sin^4 \theta \sin^2 \phi \cos \phi \\
&+ 2 \sin \theta_1 \cos \theta_1 \cos \phi_1 \sin^3 \theta \cos \theta \sin \phi \cos \phi + \sin^2 \theta_1 \sin^2 \phi_1 \sin^4 \theta \sin^3 \phi \\
&+ 2 \sin \theta_1 \cos \theta_1 \sin \phi_1 \sin^3 \theta \cos \theta \sin^2 \phi + \cos^2 \theta_1 \sin^2 \theta \cos^2 \theta \sin \phi) \\
&+ z_1^3 (\sin^3 \theta_1 \cos^3 \phi_1 \sin^5 \theta \sin \phi \cos^3 \phi + 3 \sin^3 \theta_1 \sin \phi_1 \cos^2 \phi_1 \sin^5 \theta \sin^2 \phi \cos^2 \phi \\
&+ 3 \sin^2 \theta_1 \cos \theta_1 \cos^2 \phi_1 \sin^4 \theta \cos \theta \sin \phi \cos^2 \phi + 3 \sin^3 \theta_1 \sin^2 \phi_1 \cos \phi_1 \sin^5 \theta \sin^3 \phi \cos \phi \\
&+ 6 \sin^2 \theta_1 \cos \theta_1 \sin \phi_1 \cos \phi_1 \sin^4 \theta \cos \theta \sin^2 \phi \cos \phi + 3 \sin \theta_1 \cos^2 \theta_1 \cos \phi_1 \sin^3 \theta \cos^2 \theta \sin \phi \cos \phi \\
&+ \sin^3 \theta_1 \sin^3 \phi_1 \sin^5 \theta \sin^4 \phi + 3 \sin^2 \theta_1 \cos \theta_1 \sin^2 \phi_1 \sin^4 \theta \cos \theta \sin^3 \phi \\
&+ 3 \sin \theta_1 \cos^2 \theta_1 \sin \phi_1 \sin^3 \theta \cos^2 \theta \sin^2 \phi + \cos^3 \theta_1 \sin^2 \theta \cos^3 \theta \sin \phi)) d\theta d\phi \\
&= \left(4\pi y_1^2 z_1 + \frac{4\pi z_1^3}{5} \right) \sin \theta_1 \sin \phi_1, \tag{33}
\end{aligned}$$

and

$$\begin{aligned}
I_8 &= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} x_1^3 \sin \theta \cos \theta d\theta d\phi \\
&= \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} (y_1^3 \sin \theta \cos \theta \\
&\quad + 3y_1^2 z_1 (\sin \theta_1 \cos \phi_1 \sin^2 \theta \cos \theta \cos \phi + \sin \theta_1 \sin \phi_1 \sin^2 \theta \cos \theta \sin \phi + \cos \theta_1 \sin \theta \cos^2 \theta) \\
&\quad + 3y_1 z_1^2 (\sin^2 \theta_1 \cos^2 \phi_1 \sin^3 \theta \cos \theta \cos^2 \phi + 2 \sin^2 \theta_1 \sin \phi_1 \cos \phi_1 \sin^3 \theta \cos \theta \sin \phi \cos \phi \\
&\quad + 2 \sin \theta_1 \cos \theta_1 \cos \phi_1 \sin^2 \theta \cos^2 \theta \cos \phi + \sin^2 \theta_1 \sin^2 \phi_1 \sin^3 \theta \cos \theta \sin^2 \phi \\
&\quad + 2 \sin \theta_1 \cos \theta_1 \sin \phi_1 \sin^2 \theta \cos^2 \theta \sin \phi + \cos^2 \theta_1 \sin \theta \cos^3 \theta) \\
&\quad + z_1^3 (\sin^3 \theta_1 \cos^3 \phi_1 \sin^4 \theta \cos \theta \cos^3 \phi + 3 \sin^3 \theta_1 \sin \phi_1 \cos^2 \phi_1 \sin^4 \theta \cos \theta \sin \phi \cos^2 \phi \\
&\quad + 3 \sin^2 \theta_1 \cos \theta_1 \cos^2 \phi_1 \sin^3 \theta \cos^2 \theta \cos^2 \phi + 3 \sin^3 \theta_1 \sin^2 \phi_1 \cos \phi_1 \sin^4 \theta \cos \theta \sin^2 \phi \cos \phi \\
&\quad + 6 \sin^2 \theta_1 \cos \theta_1 \sin \phi_1 \cos \phi_1 \sin^3 \theta \cos^2 \theta \sin \phi \cos \phi + 3 \sin \theta_1 \cos^2 \theta_1 \cos \phi_1 \sin^2 \theta \cos^3 \theta \cos \phi \\
&\quad + \sin^3 \theta_1 \sin^3 \phi_1 \sin^4 \theta \cos \theta \sin^3 \phi + 3 \sin^2 \theta_1 \cos \theta_1 \sin^2 \phi_1 \sin^3 \theta \cos^2 \theta \sin^2 \phi \\
&\quad + 3 \sin \theta_1 \cos^2 \theta_1 \sin \phi_1 \sin^2 \theta \cos^3 \theta \sin \phi + \cos^3 \theta_1 \sin \theta \cos^4 \theta)) d\theta d\phi \\
&= \left(4\pi y_1^2 z_1 + \frac{4\pi z_1^3}{5} \right) \cos \theta_1. \tag{34}
\end{aligned}$$

This gives a polarization

$$\begin{aligned}
\mathbf{P} &= \frac{(I_2 - I_6, I_3 - I_7, I_4 - I_8) + O(\{y_1, z_1\}^4)}{I_1 - I_5 + O(\{y_1, z_1\}^4)} \\
&= \frac{(10y_1 z_1 - 15y_1^2 z_1 - 3z_1^3)(\sin \theta_1 \cos \phi_1, \sin \theta_1 \sin \phi_1, \cos \theta_1) + O(\{y_1, z_1\}^4)}{15y_1^2 + 5z_1^2 - 15y_1 z_1^2 + O(\{y_1, z_1\}^4)} \\
&= \frac{(10y_1 z_1 - 15y_1^2 z_1 - 3z_1^3)\hat{\mathbf{B}}_1 + O(\{y_1, z_1\}^4)}{15y_1^2 + 5z_1^2} \left(1 - \frac{5y_1 z_1^2}{3y_1^2 + z_1^2} + O(\{y_1, z_1\}^2) \right)^{-1} \\
&= \frac{2y_1 z_1 \hat{\mathbf{B}}_1}{3y_1^2 + z_1^2} + \frac{(36y_1^2 z_1^3 - 45y_1^4 z_1 - 3z_1^5)\hat{\mathbf{B}}_1}{45y_1^4 + 30y_1^2 z_1^2 + 5z_1^4} + O(\{y_1, z_1\}^2), \tag{35}
\end{aligned}$$

where $\hat{\mathbf{B}}_1$ is a unit vector, in the direction of the magnetic flux density in region 1.

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